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# Viscosity and mutual diffusion in strongly asymmetric plasma mixtures

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## Abstract

We present molecular dynamics simulation results for the viscosity and mutual diffusion constant of a strongly asymmetric two-component plasma (TCP). We compare the results with available theoretical models previously tested for much smaller asymmetries. For the case of viscosity we propose a new predictive framework based on the linear mixing rule, while for mutual diffusion we point out some consistency problems of widely used Boltzmann equation based models.

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The important advancements that occurred in the last decade in the experimental techniques involving high-power lasers have generated a renewed interest in the properties of dense plasmas in general and their transport properties in particular [1–3]. The experimental capabilities currently available and the ones that are expected to become available in the near future [4] promise to further advance the field of inertial confinement fusion (ICF) as well as shed new light on long-standing astrophysics problems. Many times such experiments either probe, or their outcome is strongly dependent on, the behavior of plasma mixtures with various degrees of charge and mass asymmetries of the components. Such mixtures occur for example in ICF due to the instability (e.g. Richtmyer-Meshkov or Rayleigh-Taylor) driven mixing of the heavy elements that make up the enclosing shell and the much lighter fuel. In this case the stability of the initial interfaces, nature of the ensuing flows and degree of fuel contamination are crucially linked to such mixture properties as shear viscosity and mutual diffusion [5–7]. In the present paper we calculate these properties using molecular dynamics simulations for a simple but relevant model: the two-component plasma (TCP) [8], also known as the binary ionic mixture (BIM), which is a generalization of the one-component plasma (OCP) [9]. We study a rather extreme, ICF-inspired system [7],  $D^+ - Au^{39+}$ , which displays roughly two orders of magnitude charge and mass asymmetry, but the results should apply to other plasma mixtures as well, e.g. of astrophysics significance [10], where large charge and mass asymmetries are present. We compare the MD results with available theoretical models previously tested for much smaller asymmetries. For the case of viscosity we propose a new predictive framework based on the linear mixing rule, while for mutual diffusion we point out some consistency problems of Boltzmann equation based models.

The TCP model consists of a mixture of  $N_1$  point ions of charge  $q_1 = Z_1 e$  and mass  $M_1$  and  $N_2$  point ions of charge  $q_2 = Z_2 e$  and mass  $M_2$  embedded in a uniform, rigid, neutralizing electronic background. We denote the number concentrations by  $x_\alpha = N_\alpha/N$ ,  $N = N_1 + N_2$ ,  $\alpha = 1, 2$  and number densities by  $\rho_\alpha = N_\alpha/V$ ,  $\rho = \rho_1 + \rho_2$ , where  $V = L^3$  is the volume.  $\langle Z \rangle = x_1 Z_1 + x_2 Z_2$  is the average charge and  $\rho' = \rho_1 Z_1 + \rho_2 Z_2$  the electronic number density. As usual the mean inter-electronic and inter-ionic distances are defined by  $a' = (3/4\pi\rho')^{\frac{1}{3}}$  and  $a = (3/4\pi\rho)^{\frac{1}{3}} = a'\langle Z \rangle^{\frac{1}{3}}$ , while the electronic and ionic coupling parameters are:

$$\Gamma' = \frac{e^2}{a' k_B T} = \Gamma \langle Z \rangle^{\frac{1}{3}} \quad (1)$$

$$\Gamma_i = \Gamma' \langle Z^{\frac{5}{3}} \rangle \quad (2)$$

with

$$\Gamma = \frac{e^2}{ak_B T} \quad (3)$$

The thermodynamics of the system is fully determined by one coupling constant, e.g.  $\Gamma$ , and concentration  $x_1$ . As it is the case for the OCP, the validity range of the TCP model is such that the Fermi temperature of the electrons is  $T_F \gg T$  and the densities are high enough so  $r_s \ll 1$ ,  $r_s = at/a_0$ ,  $a_0$  - Bohr radius, corresponding to a completely degenerate and rigid electronic background.

The thermodynamics of the TCP has been thoroughly studied and is known to be very well described by the linear mixing rule [8, 11, 12]. For moderate charge asymmetries an OCP-based “one-fluid theory” is also a reasonable approximation [8], with an effective charge  $Z_{eff}^2 = \langle Z^{\frac{5}{3}} \rangle \langle Z \rangle^{\frac{1}{3}}$  suggested by the ion-sphere model [13]. The relative success of this “one-fluid” representation has lead Clerouin et al. to propose that the shear viscosity can also be predicted in terms of the equivalent OCP, as already tested for thermal transport [14]. (Although not explicitly stated in [2], further assumptions need to be made for such a prediction - see below). This “one-fluid” approach was shown to be suitable for calculating the TCP viscosity at charge and mass asymmetries of order  $\approx 10$ . Before testing this idea on much larger asymmetries,  $\approx 100$ , we note that, surprisingly, the viscosity of the OCP itself does not appear to be very accurately known. For intermediate and strong couplings,  $1 \leq \Gamma \leq 100$ , Bernu and Vieillefosse [15] have proposed an interpolation formula based on three MD simulation results obtained with systems of 108 particles. In [2] the authors propose a different relation based on the kinetic theory of Wallenborn and Baus [16], that extends to the weak coupling regime. However, the disagreement between these two approaches is significant in the regime that they both cover,  $\Gamma \geq 1$ , particularly at intermediate  $\Gamma$ 's. Unfortunately it is difficult to ascertain the reliability of these predictions given both the limited simulation results available and the small system size used, which limits the accuracy of the results.

To settle this question we performed extensive microcanonical MD simulations of the OCP with much larger system sizes - 1372 particles, and a wide range of coupling constants,  $0.05 \leq \Gamma \leq 100$ . The Coulomb interactions were handled using the Ewald summation technique with conducting boundary conditions. The calculation of the shear viscosity  $\eta$

was done using the Green-Kubo relation:

$$\eta = \frac{1}{Vk_B T} \int_0^\infty \langle \hat{\sigma}_{xy}(t) \hat{\sigma}_{xy}(0) \rangle dt \quad (4)$$

As shown by Bernu and Vieillefosse [15], and more recently in the context of Yukawa plasmas by Salin and Caillol [17], the evaluation of the pressure tensor  $\hat{\sigma}$  requires an Ewald-type summation for its interaction part,  $\hat{\sigma}^I$ :

$$\hat{\sigma} = \hat{\sigma}^K + \hat{\sigma}^I \quad (5)$$

$$\hat{\sigma}_{ab}^K = \sum_i M_i v_{i,a} v_{i,b} \quad (6)$$

$$\hat{\sigma}^I = \hat{\sigma}^{(r)} + \hat{\sigma}^{(k)} \quad (7)$$

$$\hat{\sigma}_{ab}^{(r)} = \frac{1}{2} \sum_{i \neq j} q_i q_j \frac{r_{ij,a} r_{ij,b}}{r_{ij}} \left[ \frac{2\alpha e^{-\alpha^2 r_{ij}^2}}{\sqrt{\pi} r_{ij}} + \frac{\text{erfc}(\alpha r_{ij})}{r_{ij}^2} \right] \quad (8)$$

$$\hat{\sigma}_{ab}^{(k)} = \frac{2\pi}{L^3} \sum_{|\mathbf{k}| \neq 0} \frac{e^{-\frac{k^2}{4\alpha^2}}}{k^2} \left[ \delta_{ab} - 2 \left( 1 + \frac{k^2}{4\alpha^2} \right) \frac{k_a k_b}{k^2} \right] |\tilde{\rho}(\mathbf{k})|^2 \quad (9)$$

$$\tilde{\rho}(\mathbf{k}) = \sum_i q_i e^{-i\mathbf{k} \cdot \mathbf{r}_i} \quad (10)$$

where  $a$  and  $b$  denote the Cartesian coordinates. The Ewald parameter  $\alpha$  [18] was chosen such that the real space sums, e.g.  $\hat{\sigma}^{(r)}$ , can be calculated with the usual minimum-image convention, as shown above. The duration of the runs was  $10^3 - 10^4 \omega_p^{-1}$ , where  $\omega_p^2 = 4\pi\rho e^2/M$  is the plasma frequency ( $Z = 1$ ). The natural unit for the viscosity of the OCP is  $\rho M a^2 \omega_p$ .

Our simulation results are shown in Fig. 1, together with the interpolation formula of Bernu and Vieillefosse (for  $\Gamma \geq 1$ ) and the relation proposed in [2] based on OCP kinetic theory. The present results largely agree with those of [15], but suggest that in the intermediate coupling range the OCP viscosity is significantly higher than previously predicted. The well known viscosity minimum appears to be around  $\Gamma \simeq 21$ , with  $\eta/\eta_0 \simeq 0.084$ . We are not aware of other simulations for weakly coupled plasmas,  $\Gamma \leq 1$ , but our viscosity results in this regime are in qualitative agreement with the kinetic theory of Wallenborn and Baus, although somewhat lower. Since for these conditions the screened, Debye-Hückel potential  $e^2 \exp(-r/\lambda_D)/r$ ,  $\lambda_D = [k_B T / 8\pi e^2 \rho']^{\frac{1}{2}}$ , should be an appropriate representation of the effective inter-ionic interaction and  $a/\lambda_D \propto \Gamma^{\frac{1}{2}}$ , i.e. the interaction is short ranged for  $\Gamma \ll 1$ , a good viscosity estimate should be provided by the Chapman-Enskog theory,  $\eta = 5k_B T / 8\Omega_2^{(2)}$  [19]. As shown in Fig. 1 this is indeed the case, particularly for  $\Gamma \leq 0.2$ .

We choose to fit all the data points with the relation:

$$\frac{\eta}{\eta_0} = A\Gamma^{-2} + B\Gamma^{-s} + C\Gamma \quad (11)$$

which captures rather well the behavior of the OCP viscosity in the wide range of couplings simulated. The best parameters are:  $A = 0.482$ ,  $B = 0.629$ ,  $C = 1.88 \times 10^{-3}$ , and  $s = 0.878$ .

We now tackle the question of the viscosity of a plasma mixture, in particular when the charge and mass asymmetries are very large. It should be noted that the system that we focus on,  $D^+ - Au^{39+}$ , is not a simple TCP *per se*, as gold ( $Au$ ) is only partially ionized and the effect of the remaining electrons may be important under certain thermodynamic conditions. However, the TCP approach is still relevant provided the densities and temperatures are such that the distance of closest approach between ions is larger than the radius of the remaining ion cores.

To elucidate the effect on viscosity of mixing plasmas with such large differences in charge and mass as  $D^+$  and  $Au^{39+}$  it is convenient to adopt the procedure of Ref. [2], where the coupling constant  $\Gamma$  is kept fixed and the concentration of the two species is varied. We set  $\Gamma = 0.05$ , which corresponds to  $\Gamma_i = 0.05$  for pure deuterium ( $x_{Au} = 0$ ), i.e. a weakly coupled plasma, and to  $\Gamma_i \simeq 76$  for pure gold ( $x_{Au} = 1$ ), i.e. a strongly coupled plasma. As before, we perform microcanonical simulations with a system of 1372 particles, at a number of different concentrations  $x_{Au}$ . Due to the strong charge and mass asymmetries, exceedingly long run times are necessary for both equilibration and data accumulation to calculate the viscosity using Eq. 4 with an accuracy of 20 – 25%. A good measure of the large “size” difference between the ions is provided for example by the pair correlation functions, which we show in Fig. 2 for  $x_{Au} = 0.5$ .

As noted in [2] the viscosity drops steeply upon mixing highly charged, heavy ions in a weakly coupled plasma, see Fig. 3. This effect can be understood qualitatively in the framework of a “one-fluid” theory, which we describe below. The coupling constant of the equivalent OCP is  $\Gamma_{eff} = \Gamma Z_{eff}^2$ , where  $Z_{eff}^2 = \langle Z^{\frac{5}{3}} \rangle \langle Z \rangle^{\frac{1}{3}}$  follows from the ion-sphere model [13]. In this approximation the thermodynamics is fully determined by  $\Gamma_{eff}$ , but the calculation of the viscosity requires some additional arguments. For example, a reasonable unit for the viscosity of this system may be assumed to be  $\eta_0^m = \rho \langle M \rangle a^2 \omega_{pm}$ ,  $\langle M \rangle = x_1 M_1 + x_2 M_2$ , where  $\omega_{pm}^2 = \omega_p^2 \langle Z \rangle^2 / \langle M \rangle$  is the “hydrodynamic” plasma mixture frequency. (The use of the so-called “kinetic” mixture frequency [20] leaves the results largely unchanged). The

mixture viscosity in these units is then postulated to be given by the scaled effective-OCP viscosity:

$$\frac{\eta(\Gamma, x)}{\eta_0^m(x)} = \frac{\eta_{OCP}(\Gamma_{eff})}{\eta_0} \quad (12)$$

We show in Fig. 3 the results of such calculations for the system that we study, using both the OCP viscosity of Ref. [2], and our prediction Eq. 11. The qualitative dependence of the MD results on  $x_{Au}$ , arising from the opposite behaviors of  $\eta_{OCP}(\Gamma_{eff})$  and  $\eta_0^m(x)$ , is reproduced correctly, but the quantitative disagreement is also very significant, particularly for small and moderate  $Au$  concentrations. The use of the more accurate OCP viscosity Eq. 11 does not fully alleviate this problem. We conclude that, not surprisingly, the accuracy of the “one-fluid” model is diminished for extreme asymmetries.

It is clear that this limitation can only be overcome by taking into account, either explicitly or implicitly, the mixture asymmetry. A direct calculation along the lines of the kinetic theory of Wallenborn and Baus [16] has been used for example to determine the TCP mutual diffusion constant at small asymmetries [21]. However, the calculation of the viscosity is even more complex and given the limitations of the theory even for the OCP its success for TCP quantitative predictions is rather doubtful. We turn therefore to a more indirect method, which we outline below. First, we recall an interesting and much studied property of two-component plasmas, the linear mixing rule [8, 11, 12]. Hansen et al. have pointed out that the excess internal energy of the TCP,  $u = U_{ex}/Nk_B T$ , is very accurately represented as a linear combination of the excess energies of two one-component plasmas with the same electronic coupling constant  $\Gamma'$  as the TCP (and ionic charges  $Z_1 e$  and  $Z_2 e$ ), i.e. the mixing of the two components at the same temperature and electronic density is largely ideal:

$$u(\Gamma', x_1) \simeq x_1 u_{OCP}(\Gamma' Z_1^{\frac{5}{3}}) + x_2 u_{OCP}(\Gamma' Z_2^{\frac{5}{3}}) \quad (13)$$

We find that this rule is satisfied at very large asymmetries as well, with the largest relative deviations occurring at  $\Gamma' \ll 1$ , i.e. small  $Au$  concentrations, in agreement with [8]. Given this nearly ideal mixing behavior we assume that other system properties, e.g. viscosity, are bracketed by the component values as well. For an interpolation relation between the viscosities of the two OCP’s at a given composition we now borrow some concepts from the linear transport theory of composite media. A common situation encountered in such



systems is that of “impurities” with a generalized conductivity  $\alpha_1$  and total volume fraction  $\phi_1$  randomly dispersed in a matrix  $\alpha_2$ . Under these circumstances the effective medium theory [22] employs a mean-field like, self-consistent approximation to predict the medium conductivity on scales much larger than those of the inhomogeneities. For the case of viscosity this yields [23] for the effective medium viscosity  $\eta_m$ :

$$\sum_i \phi_i \frac{\eta_i - \eta_m}{\eta_i + \frac{3}{2}\eta_m} = 0 \quad (14)$$

We now note that the TCP is obtained by combining one-component plasmas with volume fractions  $\phi_i = Z_i x_i / \langle Z \rangle$  and use the above relation to predict the mixture viscosity, essentially assuming that the theory applies for atomically sized “impurity” domains. Using Eq. 11 for the individual OCP viscosities  $\eta_1$  and  $\eta_2$  we obtain the results shown in Fig. 3.

The degree of agreement with the simulation results is fairly remarkable. This may lead one to believe that the system is perhaps thermodynamically unstable and separating into two OCP phases [8]. However, we find no evidence for such a scenario and conclude that the decoupling signaled by the linear mixing rule along with the tremendous asymmetry between the ions lead to behavior mimicking that of a macroscopically mixed system.

We now turn to the case of ionic interdiffusion. Mutual diffusion in plasma mixtures plays an important role in the prediction of stellar structure [24], as well as the stability of ICF targets [5]. For the case of a binary mixture the mutual diffusion coefficient  $D_{12}$  can be calculated in terms of the fluctuations of the microscopic interdiffusion current [25]:

$$D_{12} = x_1 x_2 \left[ \frac{\partial^2 (\beta G / N)}{\partial x_1^2} \right]_{P,T} D_{12}^0 \quad (15)$$

$$D_{12}^0 = \frac{1}{3N x_1 x_2} \int_0^\infty \langle \mathbf{j}_c(t) \mathbf{j}_c(0) \rangle dt \quad (16)$$

$$\mathbf{j}_c(t) = x_2 \sum_1^{N_1} \mathbf{v}_i(t) - x_1 \sum_1^{N_2} \mathbf{v}_i(t) \quad (17)$$

$$(18)$$

where  $G$  is the Gibbs free energy. The thermodynamic factor that multiplies the Green-Kubo component  $D_{12}^0$  reduces to unity for dilute gas mixtures [25], but in low density, weakly coupled plasmas goes to  $\langle Z^2 \rangle / \langle Z \rangle^2$ , which has been interpreted as an effect of the ambipolar electric field of the electrons [21]. It is worth noting that the above relation is a good estimate for  $x_1 x_2 [\partial^2 (\beta G / N) / \partial x_1^2]_{P,T}$  at weak as well as strong couplings. Since the

linear mixing rule holds well for all couplings, the change in Helmholtz free energy upon mixing at constant electronic density (and temperature):

$$\frac{\Delta F}{N} = f(\Gamma', x_1) - x_1 f_{OCP}(\Gamma' Z_1^{\frac{5}{3}}) - x_2 f_{OCP}(\Gamma' Z_2^{\frac{5}{3}}) \quad (19)$$

is very well approximated by the ideal entropy of mixing (with negative sign):

$$\frac{\Delta F}{N} \simeq k_B T \left[ x_1 \ln \frac{x_1 Z_1}{\langle Z \rangle} + x_2 \ln \frac{x_2 Z_2}{\langle Z \rangle} \right] \quad (20)$$

If we assume that the pressure is entirely determined by the electronic density, i.e.  $p_{electronic} \gg p_{ionic}$ , which is consistent with the initial assumption  $r_s \ll 1$ , then  $\Delta G = \Delta F$ . We can therefore immediately calculate the thermodynamic factor as  $\langle Z^2 \rangle / \langle Z \rangle^2$ . The difference between plasma and ideal gas mixtures appears here to be an entropic effect induced by the charge neutralizing background, as mixing occurs at constant electronic density (i.e. constant electronic pressure) as opposed to constant molecular density (i.e. constant ideal gas pressure).

In the course of the molecular dynamics simulations with various  $D^+ - Au^{39+}$  mixtures we also calculated the microscopic interdiffusion current  $\mathbf{j}_c$ , and therefore were able to determine the Green-Kubo integrand  $D_{12}^0$ . The results are shown in Fig. 4 relative to  $D_0 = a^2 \omega_p$ , along with the discussed thermodynamic factor estimate. We find that  $D_{12}^0$  is almost concentration independent for  $x_{Au} \geq 0.1$ , but appears to decrease fairly steeply at lower concentrations. The factor  $\langle Z^2 \rangle / \langle Z \rangle^2$  has a simple behavior, with a sharp maximum for small amounts of highly charged ions.

For small ionic asymmetries kinetic theory estimates of  $D_{12}^0$  were found to be in good agreement with simulations [21]. A simpler model, widely employed for astrophysics problems, was proposed by Paquette et al. [26]. Its main assumption is that the Boltzmann equation can be used to calculate the transport coefficients of plasma mixtures by making use of the Chapman-Enskog solution method [19]. The authors further argue that screened potentials,  $Z_i Z_j e^2 \exp(-r/\lambda)/r$ , are better suited for such estimates than the pure Coulomb interaction. In order to extend the validity of this approach to strong couplings they propose as appropriate screening distance  $\lambda$  the larger of  $\lambda_D$  and  $a$ , where  $\lambda_D$  is the Debye screening length:

$$\lambda_D = \left[ \frac{k_B T}{4\pi e^2 (\rho' + \sum_i \rho_i Z_i^2)} \right]^{\frac{1}{2}} \quad (21)$$

Under these assumptions the Boltzmann equation mutual diffusion coefficient is given in the first Enskog approximation as:

$$[D_{12}]_1 = \frac{3k_B T(M_1 + M_2)}{16\rho M_1 M_2 \Omega_{12}^{(11)}} \quad (22)$$

where  $\Omega_{12}^{(11)}$  are collision integrals [19] that have been tabulated with high accuracy in Ref. [26]. We perform such calculations for the  $D^+ - Au^{39+}$  mixture using the slightly better second Enskog approximation [26]. The outcome, see Fig. 4, reproduces rather well the MD simulation results for  $D_{12}^0$  at  $x_{Au} \geq 0.1$ , but *not* the mutual diffusion coefficient  $D_{12}$ . This is an important point that merits further discussion. In fact, there is no reason to expect that Chapman-Enskog estimates based on the Boltzmann equation for the ions can reproduce the full  $D_{12}$  for either pure Coulomb interactions (with some reasonable cut-off) or screened potentials. As shown in [21] for low density plasmas or more generally here, the enhancement factor  $\langle Z^2 \rangle / \langle Z \rangle^2$  only arises when the electronic background is explicitly taken into account either through its ambipolar field in a dilute plasma kinetic description [21] or simply at the thermodynamic level in the context of the linear mixing rule. No such effect is included when the standard Boltzmann equation, which is consistent with ideal gas thermodynamics, is used to model the dynamics of the ions. Therefore it is reasonable to expect that such approaches can only provide estimates of the Green-Kubo part, i.e.  $D_{12}^0$ , of the mutual diffusion constant, as already evidenced by our simulation results and perhaps not fully appreciated before. Furthermore, the plasma thermodynamic factor is large for large charge asymmetries particularly at small concentrations of highly-charged heavy ions, a situation quite typical in many astrophysics problems [24].

The apparent failure of the screened potentials method proposed by Paquette et al. at low  $Au$  concentrations may appear at first puzzling since as  $x_{Au}$  decreases so does the effective coupling constant  $\Gamma_{eff}$  and therefore the theory should become in principle rigorously valid. We note however that this also requires  $\lambda_D \gg a$ , which for our system and chosen  $\Gamma$  means  $x_{Au}$  no bigger than  $\approx 10^{-3}$ . In fact, although not easily seen in Fig. 4, the theoretical values drop sharply for such compositions to values close to the MD result at the lowest simulated  $Au$  concentration,  $x_{Au} = 0.03$ . For this composition the screening distance  $a$ , although larger than  $\lambda_D$ , appears to be too small. This is perhaps not too surprising given that due to the significant separation between the highly-charged ions they are primarily screened by the small ions and the electronic background, which should require distances significantly

larger than  $a$  when the charge asymmetry is very large.

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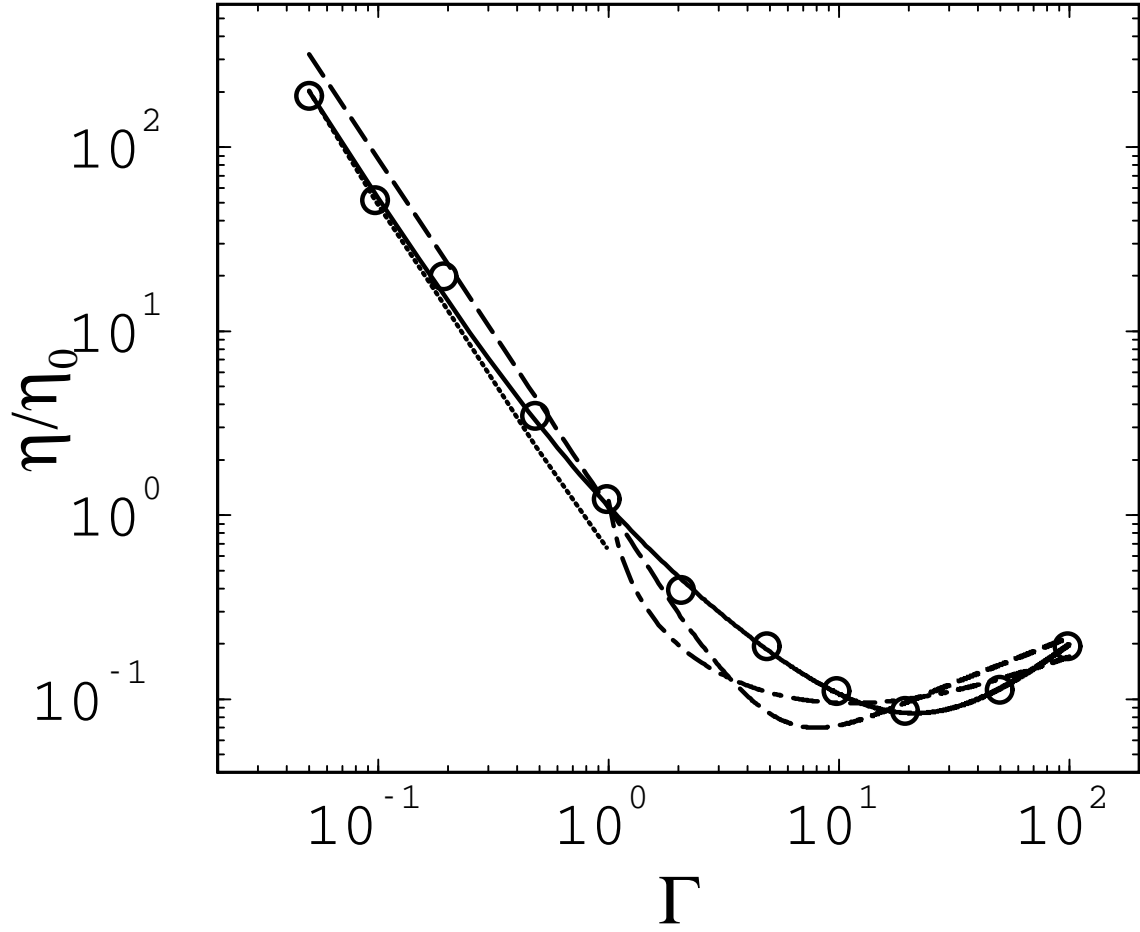


FIG. 1: OCP viscosity: present simulations (circles), fit of simulation results - Eq. 11 (solid line), predictive relation of Ref. [2] (dashed line), Bernu-Vieillefosse interpolation formula [15] (dot-dashed line), Chapman-Enskog estimate using screened potentials (dotted line).

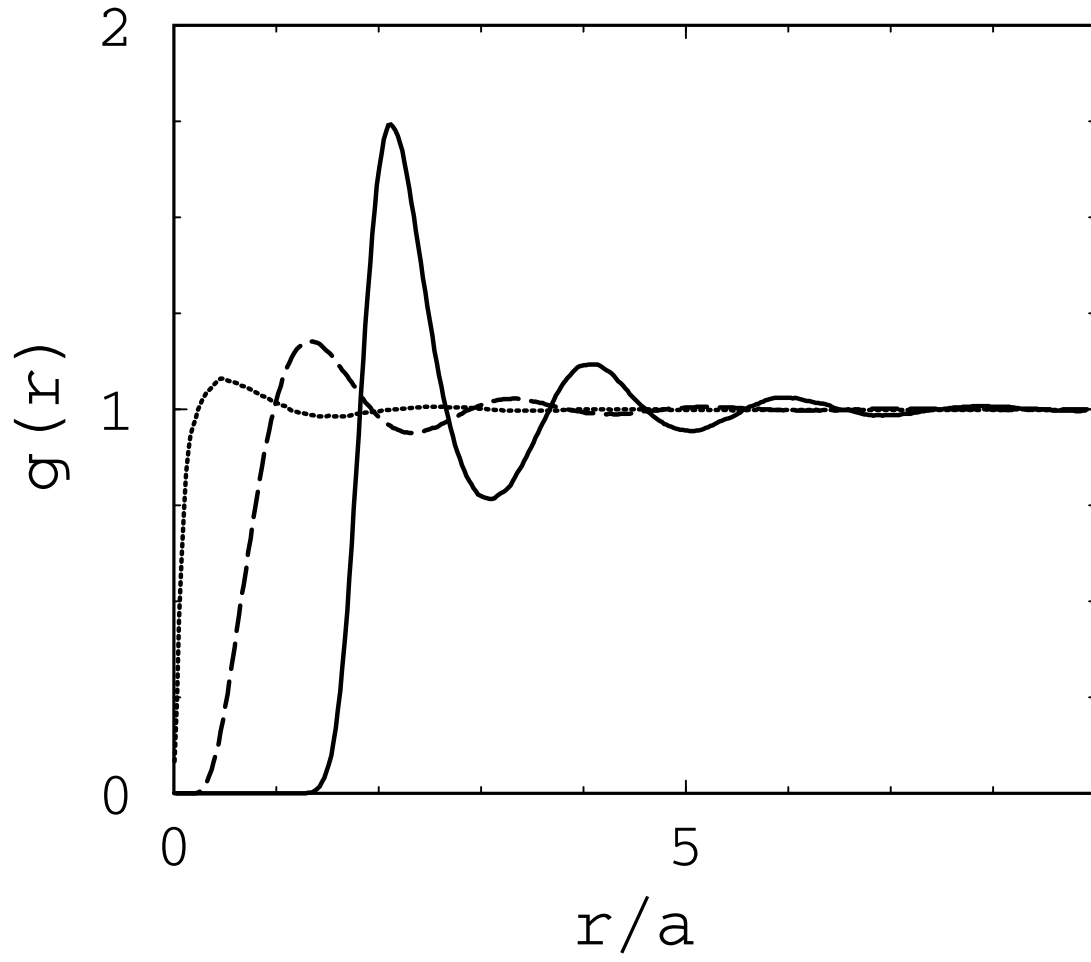


FIG. 2: Ion-ion pair correlation functions for  $x_{Au} = 0.5$ :  $D^+ - D^+$  (dotted line),  $D^+ - Au^{39+}$  (dashed line),  $Au^{39+} - Au^{39+}$  (solid line).

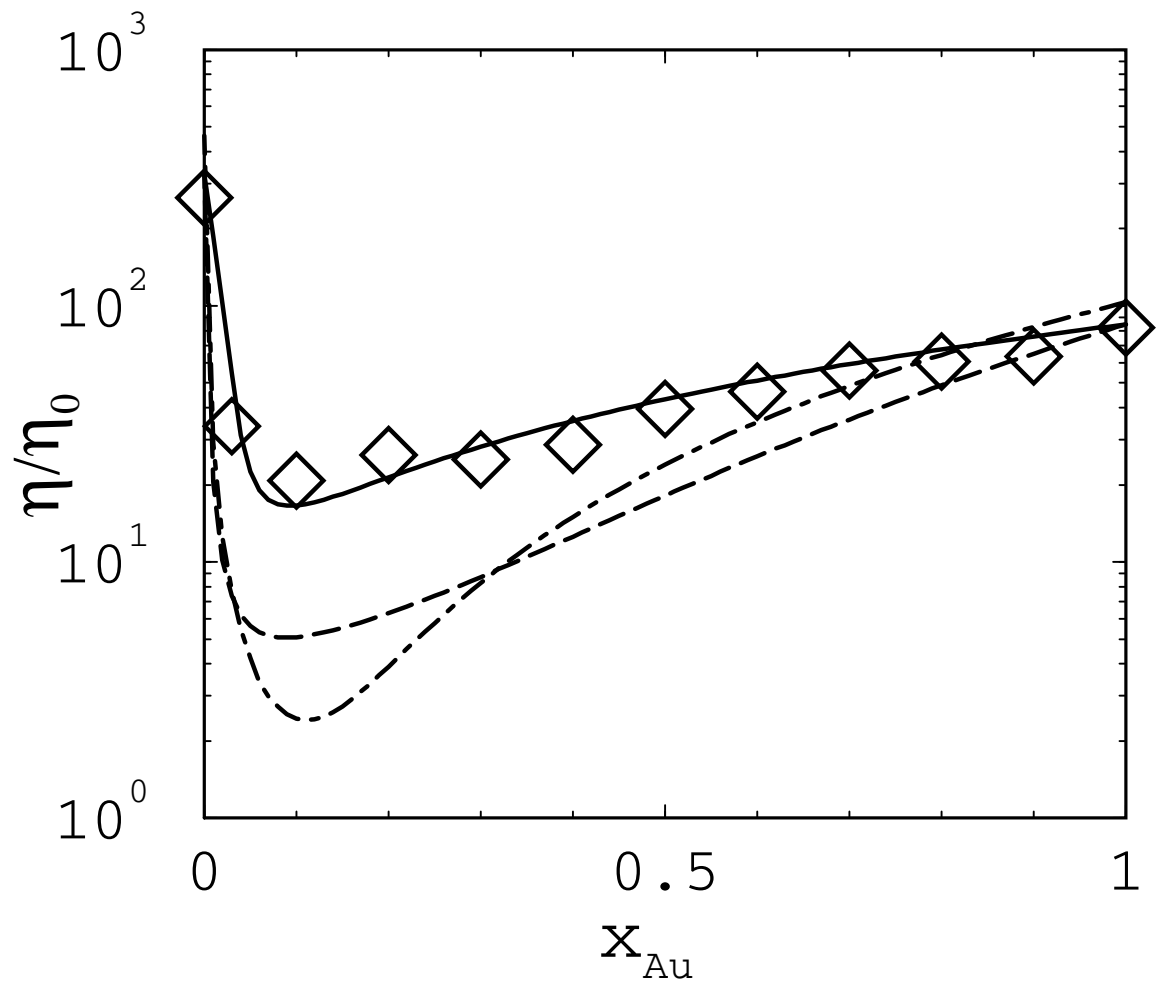


FIG. 3: Viscosity of the  $D^+ - Au^{39+}$  TCP: simulations (diamonds), one-component model of Ref. [2] (dot-dashed line), one-component model using Eq. 11 for the OCP viscosity (dashed line), two-component model Eq. 14 (solid line).



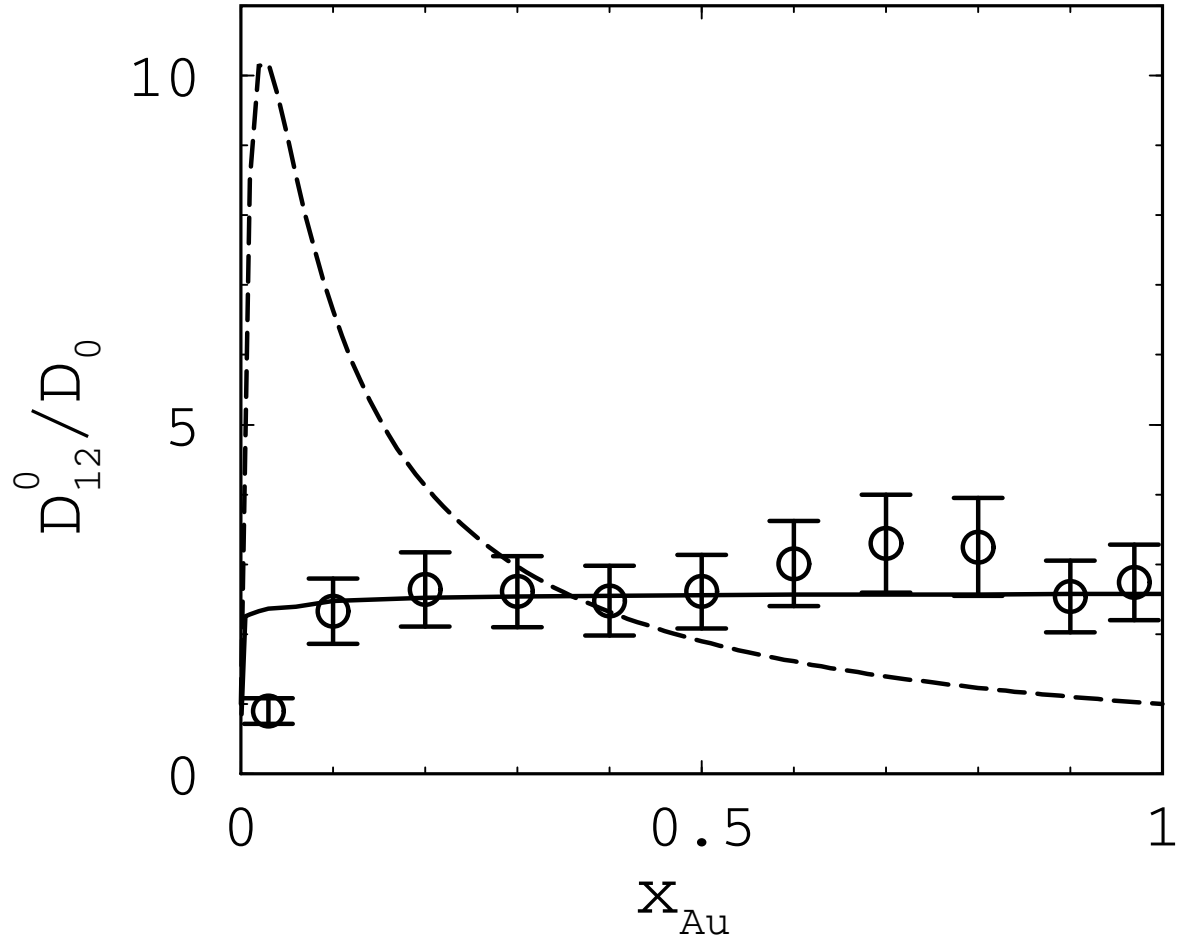


FIG. 4: Mutual diffusion constant contribution  $D_{12}^0$  of the  $D^+ - Au^{39+}$  TCP: simulations (circles), screened potential model of Ref. [26] (solid line). Thermodynamic factor  $\langle Z^2 \rangle / \langle Z \rangle^2$  (dashed line).